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JÜLICH FORSCHUNGSZENTRUM

Fuel for Gas-Cooled Reactors, mainly HTR

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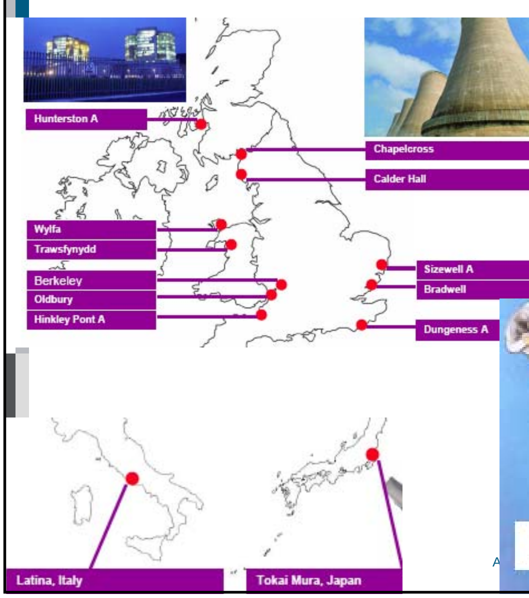
1 Gas-Cooled Reactors

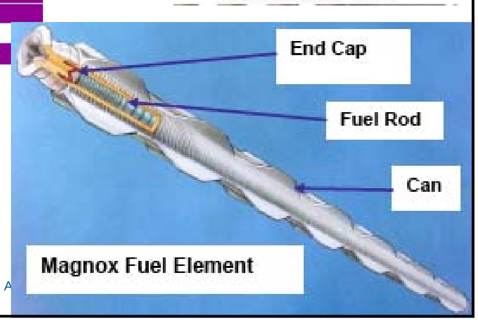
Fermi pile➔Magnox➔AGR➔HTR

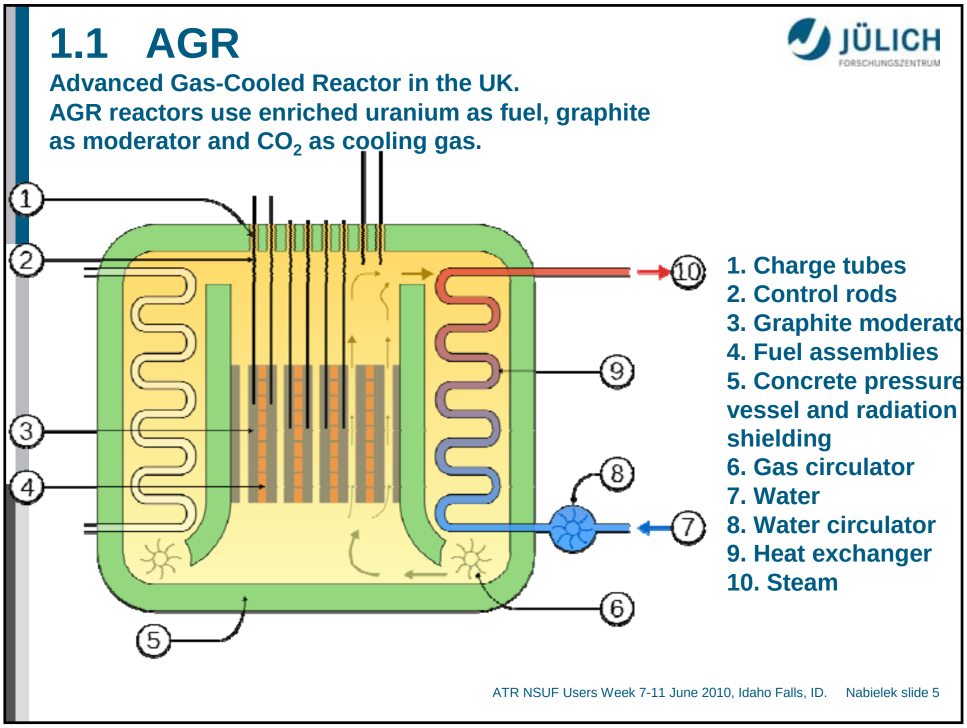
various names: HTR, HTGR, MHTGR, PBMR, GT-MHR

1 Magnox



Fuel rod insertion 1950's





1.1 AGR:

Advanced Gas-Cooled Reactors in the UK. A total of 14 reactor units of this type are in operation in England and Scotland. The plan was that they were going to be replaced by HTRs, but this was not realized in Britain.

* Coolant: CO_2
* Moderator: Graphite
* Fuel is uranium dioxide pellets,
  enriched to 2.5-3.5%,
  in stainless steel tubes.
* Cladding: stainless steel. In graphite sleeves.
* Operating pressure: 40 atmospheres
* Coolant exit temperature: 650°C

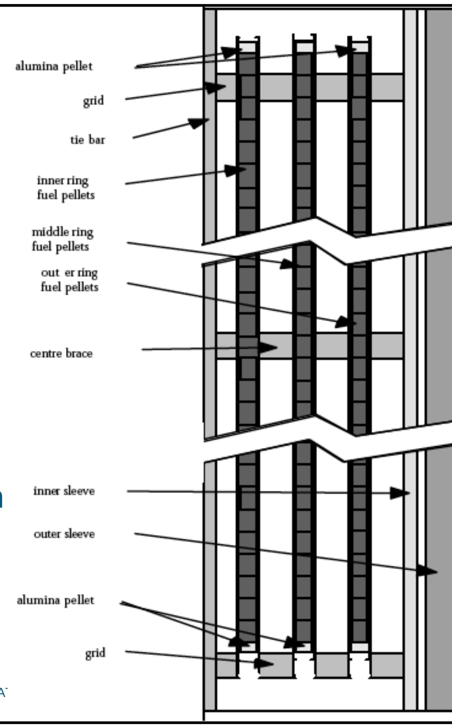
Designed for on-load refuelling,
however not licensed at full power operation

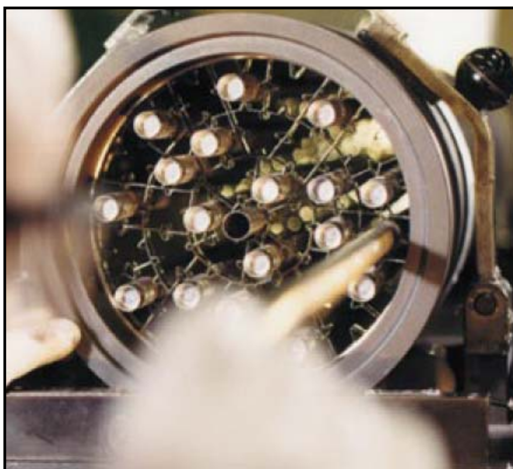
Electrical Efficiency 42%

1.1 AGR fuel:

AGRs are using UO_2 fuel pellets enriched between 2-3% encased in 14.5 mm diameter stainless steel tubes about 1m long.

There are 64 pellets per 950 mm fuel length.
The fuel elements consist of 36 fuel rods spaced within a grid framework which is enclosed within an inner and outer graphite sleeve.





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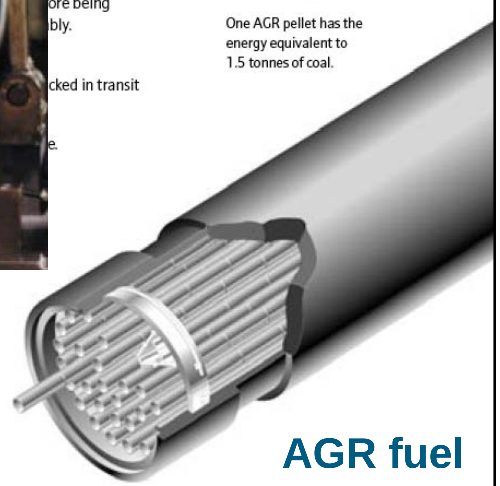
One AGR pellet has the energy equivalent to 1.5 tonnes of coal.

stainless steel fuel pins each containing 64 UO_2 fuel pellets. A single AGR assembly produces the same amount of electricity as 3,600 tonnes of coal.



AGR fuel

1.2 HTR: Ceramic Materials in GenIV Nuclear Systems are required

- High efficiency $0.4<\eta_{el}<0.5$
- Ceramic materials and helium coolant
  - avoid interactions and phase changes
  - are stable to very high temperatures
- Large heat capacity gives passive safety features to
  - provide confidence
  - simplify plant and control systems
- Many diverse applications beyond electric power
  - steam for chemical industry
  - process heat applications including H_2 production

1.2 High Temperature Reactor

- <u>700-750°C helium coolant exit temperature:</u> steam for power generation and/or steam for chemical industry
- <u>850-900°C helium coolant exit temperature :</u> direct cycle gas turbine for very high efficiency and reliability
- <u>950-1000°C helium coolant exit temperature :</u> process heat including H_2 production
- [AVR Jülich increased temperatures from 750° to 850° to 950°C in the year 1974…..]

BROAD FOUNDATION OF HELIUM REACTOR TECHNOLOGY

source: Eben Mulder Potchefstrom Univ

BASIC RESEARCH VALIDATION

EXPERIMENTAL REACTORS

DEMONSTRATION OF BASIC HTGR TECHNOLOGY

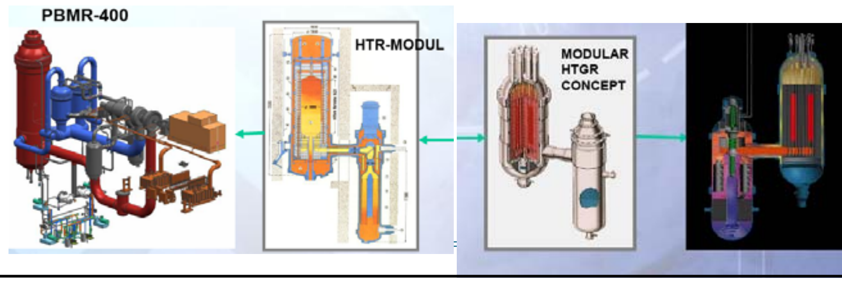
DRAGON
(U.K.)
1963 - 76

AVR
(FRG)
1967 - 1988

PEACH BOTTOM 1
(U.S.A.)
1967 - 1974

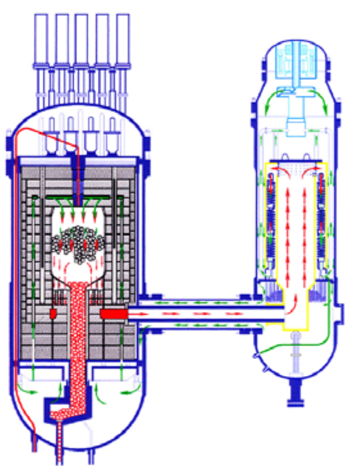
FORT ST. VRAIN
(U.S.A.)
1976 - 1989

THTR
(FRG)
1986 - 1989



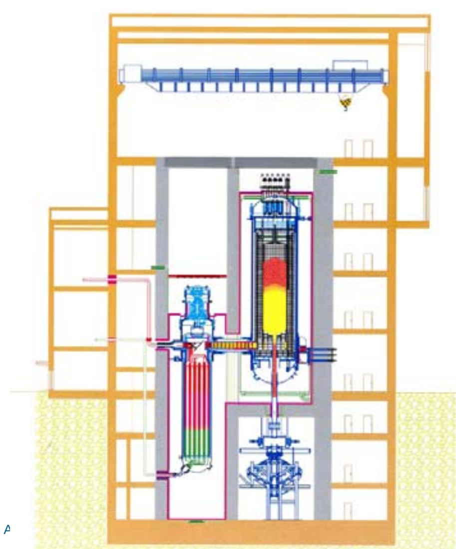
Pebble-Bed HTRs in China

HTR-10



HTR-PM

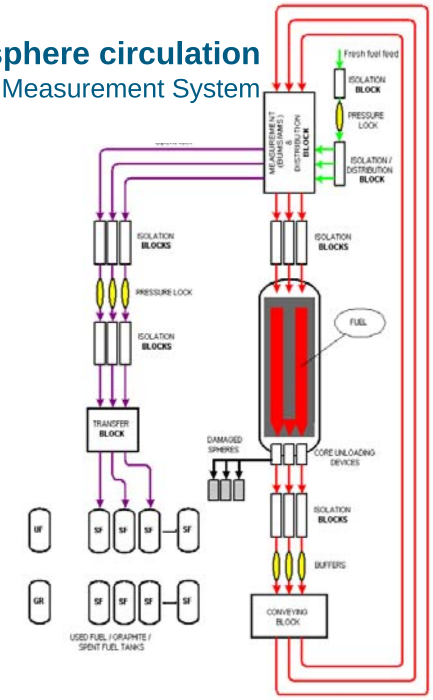
source: INET



1.2.1 Pebble-Bed BUMS and sphere circulation

BUMS = BUrnup Measurement System

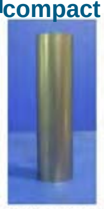
In a pebble-bed reactor, spherical fuel elements drop slowly through the core, are removed, checked for integrity, measured for burnup and re-inserted.



1.2.2 HTR Prismatic Design

source: GA

compact

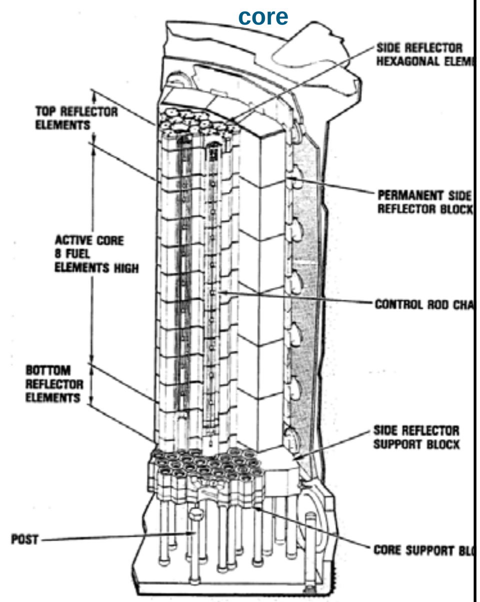


12.5 mm Diameter
~50 mm Long

block

360 mm Across Flats
~800 mm Long

core

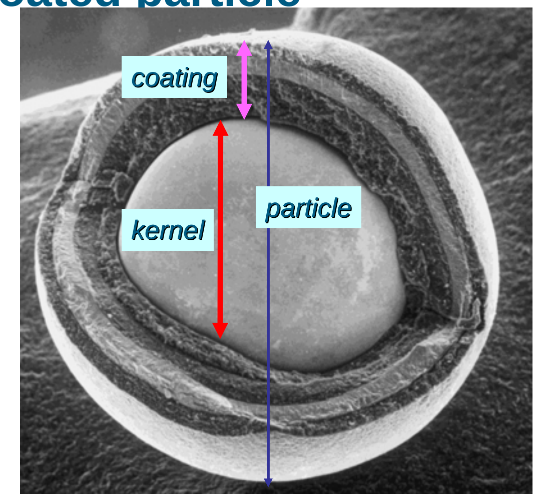


2 HTR Fuel
The ceramic coated particle

HTR
1954 P Fortescue
1956 R Schulten

History of coated particles:
1957 R A U Huddle
1959 W Goeddel
1961 J Oxley, Battelle
fluidised bed coating

Manufacturing
United States
United Kingdom
France
Belgium
Germany
Russia
India
Japan
China
South Africa
South Korea

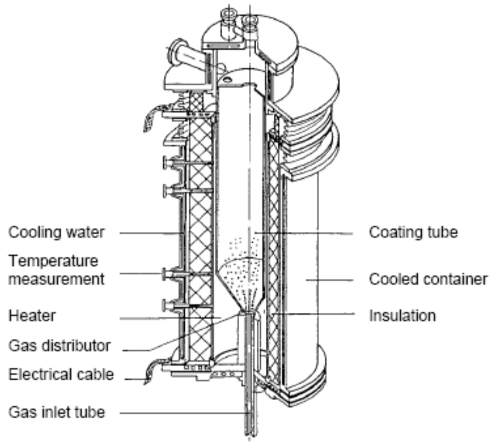


2.1 Worldwide History of HTR Fuel Fabrication

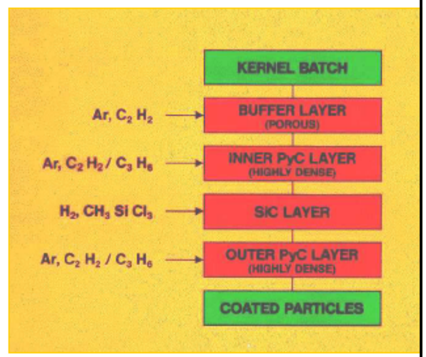
Reactor/ Manufacturer	Fuel Description	Total HM (kg)
ROVER/GA LANL	BISO in rods	1
Peach Bottom/ GA	BISO in compacts	3,500
UHTREX/GA LANL	BISO in compacts	200
DRAGON	TRISO, BISO compacts	300
FSV/ GA	TRISO in compacts	33,400
THTR/ NUKEM	BISO in spheres	11,000
AVR/ NUKEM	HEU BISO, TRISO spheres	1,700
AVR/ NUKEM	Modern LEU TRISO spheres	480
US development GA	Modern TRISO UCO	500
HTTR/ NFI	Modern LEU TRISO compacts	900
HTR-10/ INET	Modern LEU TRISO spheres	135

Historic fuel from 1964 to 2001. Recent: USA, SA, France
Fuel quality was improved from 10^{-1} to 10^{-5}

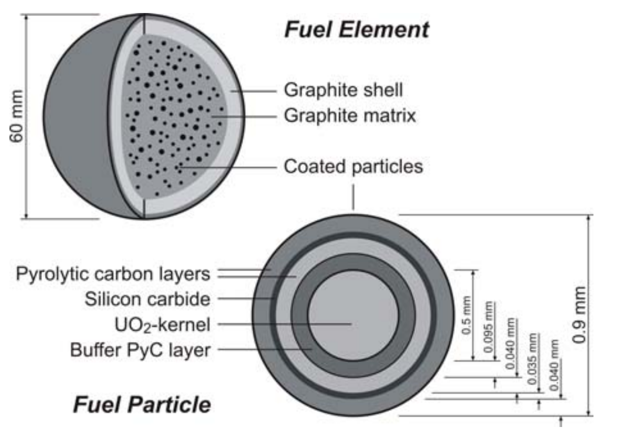
2.1 CVD coating technology



Particle coating



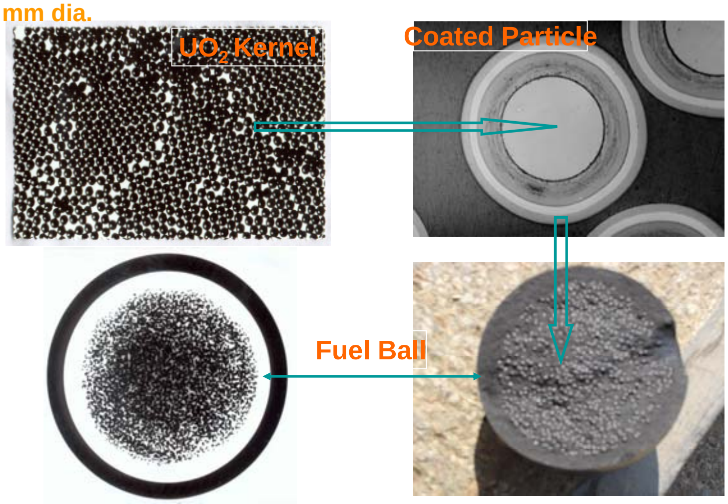
2.1 Spherical fuel element and coated particle



2.1 HTR Spherical Fuel Element

source: INET

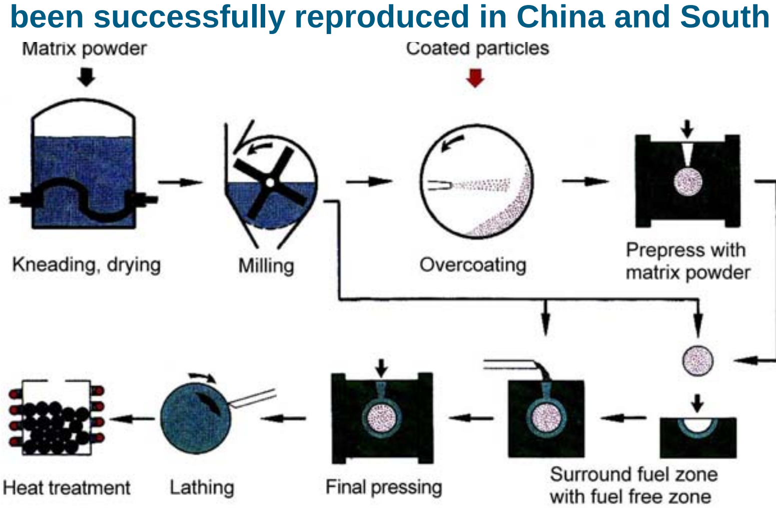
0.5 mm dia.



1 mm dia

60 mm dia.

2.1 German Manufacture of Spherical Fuel Elements has been successfully reproduced in China and South Africa

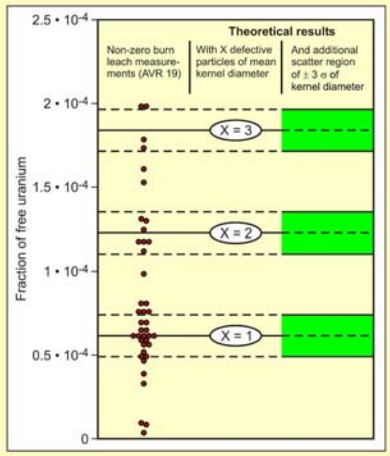


Source: NUKEM

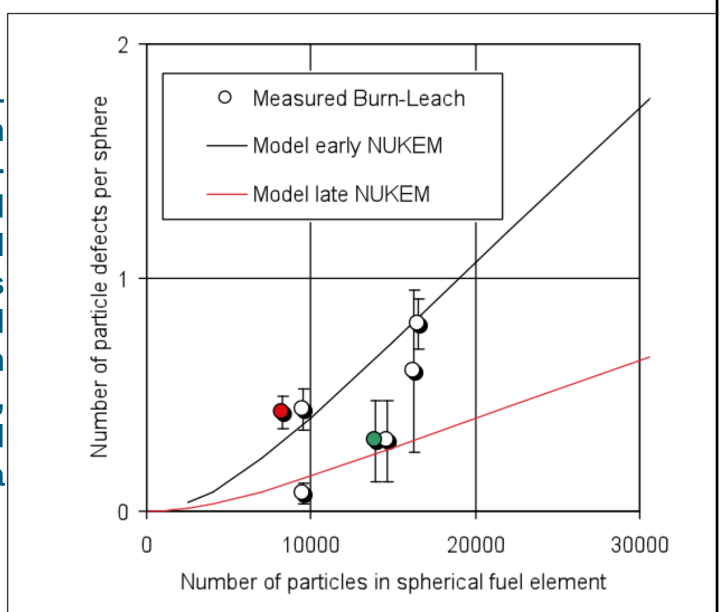
2.1 Free uranium measurements from burn-leach tests on spherical fuel elements from the AVR GLE-3 reload

Modern fuels

- With the production of LEU TRISO fuel (GLE-3 and GLE-4), the high quality level of German HTGR fuel production could be proven.
- From the approximately 24,600 GLE-3 fuel spheres (reload charge 19), 70 were taken for quality control.
- Results show that the U_{free}/U_{total} ratios were observed to be integer multiples of a coated particle inventory ($1/16400 = 0.61 \cdot 10^{-4}$), meaning that the free uranium is mainly from particles with a defective SiC coating.
- It demonstrated thus the extremely low contamination level of the matrix material with U-235 basically originating from natural contamination of the graphitic raw materials.



2.1 Burn-leach results on as-manufactured spherical fuel elements compare well between Germany, China and South Africa



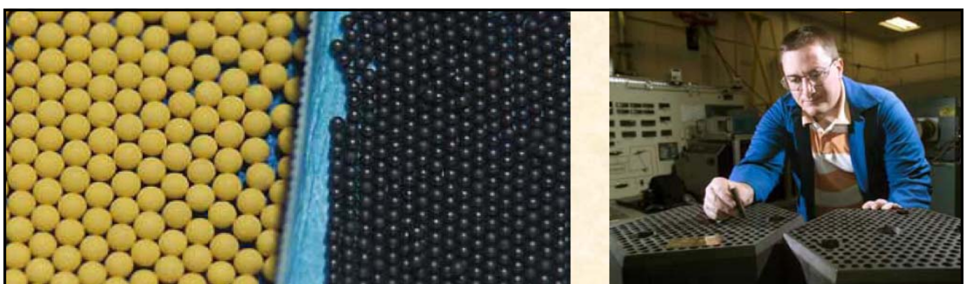
Number of particle defects in manufacture as a function of the number of particles in a spherical fuel element. The red circle are Chinese 2001 results and the green circle are African 2009 results.

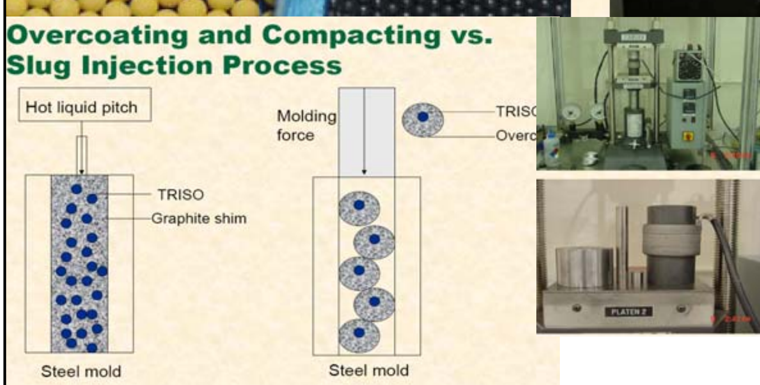
2.1 US HTR fuel manufacture 1970-1995

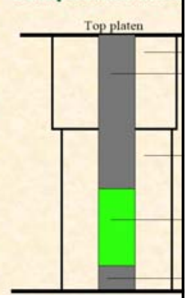
FUEL KERNEL GELATION UNIT

source: Saurwein, GA

FLUIDIZED-BED TRISO PARTICLE COATER







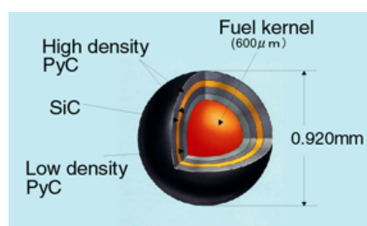
2.1 Recent US kernels, particles, compacts from 2000+. Source: Pappano ORNL

2.1 HTR
all-ceramic core materials, therefore temperatures up to 1000°C are possible

<table>
<tr><td>Moderator</td><td colspan="3" rowspan="3">Nuclear Graphite</td></tr>
<tr><td>Reflector</td></tr>
<tr><td>[Fuel Element]</td></tr>
<tr><td rowspan="4">Fuel Body</td><td colspan="3">Matrix Graphite</td></tr>
<tr><td rowspan="3">Coated Fuel Particle</td><td colspan="2">UO_2 kernel</td></tr>
<tr><td rowspan="2">Coating</td><td>Pyrocarbon</td></tr>
<tr><td>SiC</td></tr>
</table>

2.1 Coated Fuel Particle

- **Buffer Layer (Low density carbon, 95 µm)**
  - **Void volume for fission gases**
- **iPyC (40 µm)**
  - **Stop gaseous fission products and protects SiC**
  - **Seals off the kernel**
- **SiC (35 µm)**
  - **Retain gas and metal fission products**
  - **Enhances mechanical stability**
- **oPyC (40 µm)**
  - **Protects SiC mechanically**
  - **Fission product barrier in particles with defective SiC**



Coated fuel particle

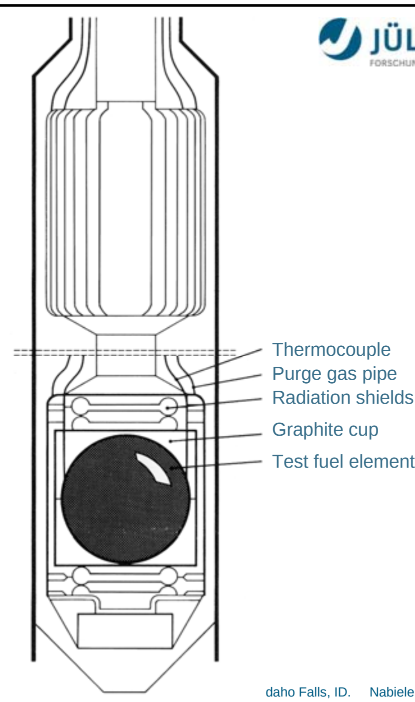
2.1 Each layer in the Triso particle design plays a role in fuel performance and fission product retention

LAYER	FUNCTION
Oxide Fuel Kernel	Contains Fuel Material Diffusion Barrier/ Chemical Holdup Structural Base of Particle
Buffer	Void Volume for Gaseous FP Accommodates Kernel Swelling Sacrificial Layer for Fission Fragments
inner Pyrocarbon	Gastight Coating/ Protects Kernel from Cl_2 Diffusion Barrier Metallic FP Reduces Tensile Stress on SiC
SiC	Primary Metallic FP Diffusion Layer Primary Pressure Retaining Layer
outer Pyrocarbon	Gaseous FP Barrier Diffusion Barrier Metallic FP Reduces Tensile Stress on SiC Provides Bonding Surface for Overcoating, or Matrix Material, respectively

2.2 Irradiation Test Rig & Capsule

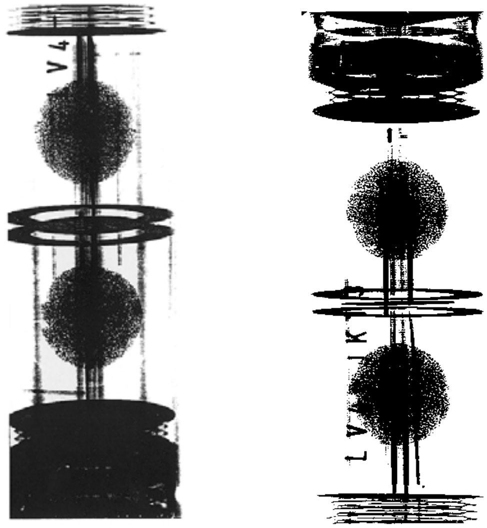
The fuel elements in an irradiation test are contained in one or more instrumented, independent capsules that provide:

- Fuel Temperature Monitoring,
- A Closed Purge Gas System for:
  - Active Temperature Control (He/Ne Ratio)
  - On-Line Fission Gas Release Monitoring
    (via quantitative gamma spectrometry)
- Dosimetry Materials (fluence)
- Neutron Flux Detectors.



Capsule 1

Capsule 2

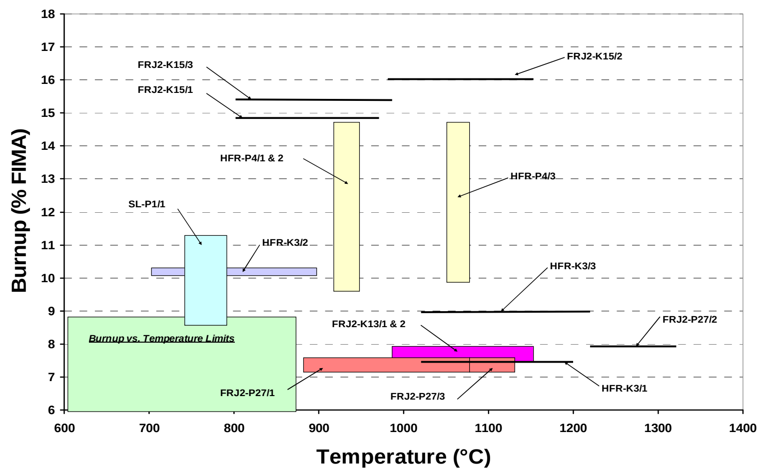


2.2 X-ray photograph from the assembled capsules 1 and 2 for MTR irradiation experiment FRJ2-K13

2.2 LEU TRISO UO_2 IRRADIATION TESTS

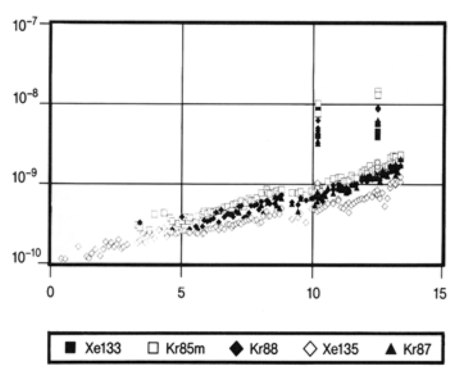
- **Phase 1 Irradiation Tests – Identify and quantify all mechanisms contributing to fission product release**
- **Phase 2 Irradiation Tests – Simulation of manufacturing process and operational conditions for HTR-Modul Reactor Design**
- **Real-Time AVR Irradiation - Bulk testing of fuel under reactor operating conditions**

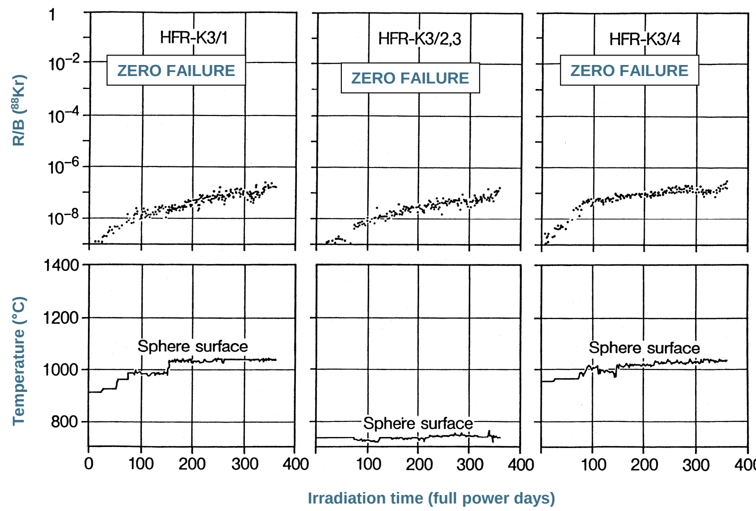
2.2 Testing range



2.2 On-Line Monitoring of Fission Gas Release

Release Rate to Birth Rate (R/B) of fission gases as a function of burnup from experiment FRJ2-K15 in the Jülich DIDO reactor . The spikes in R/B were measured during +200°C temperature transients. The slow increase in R/B is from uranium contamination of natural enrichment.





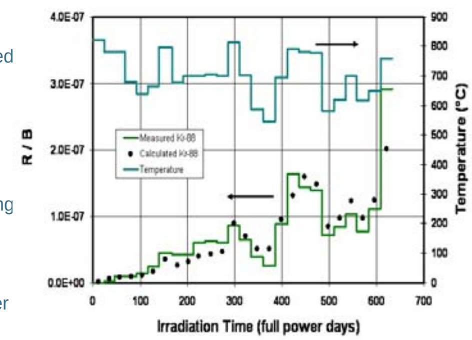
2.2 Gas release rates and temperatures during a typical irradiation of spherical fuel elements (experiment HFR-K3)

2.2 Accelerated MTR Testing & HTR Modul Proof Testing

LEU UO_2 TRISO Particle Performance

- Based on ^{85m}Kr & ^{88}Kr Noble gas release measured at EOL of all MTR experiments
- Total of 19 reference spherical elements tested and evaluated:
  - 11 elements / 4 Accelerated MTR Tests
  - 8 elements / 2 HTR Proof Tests
- No in-reactor failure observed in 278,760 particles tested to HTR Modul normal operating conditions.
- Collectively, the EOL failure level observed from reference elements was 1.07×10^{-5} (upper 95% confidence limit)
- In-reactor particle failure was observed in some non-reference fuel specimens

^{88}Kr R/B values for HFR-K6/Capsule 2 throughout its 634 day irradiation



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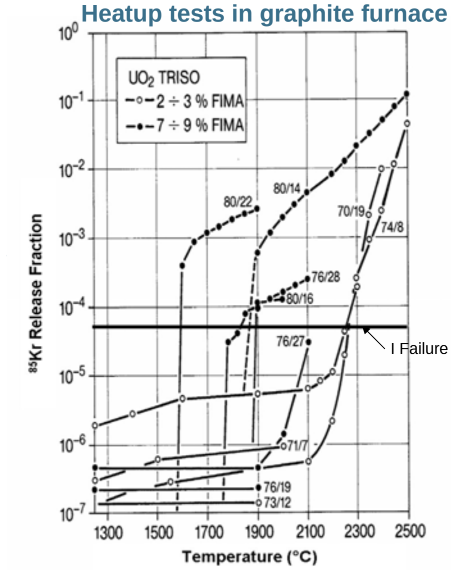
2.2 AVR Real-Time Irradiation Testing

GLE 3 Fuel Element Performance

- Based on ^{85}Kr Noble gas release measured at 1250°C during accident simulation testing
- 14 GLE Elements Investigated:
  Burnup: 1.7 to 9.25% FIMA,
  Fast Fluence: 0.19 to 2.94 x10^{25} n/m^2, E >16 fJ
  Operating Temperature: 950°C < T < 1380°C
- No failures observed in 233,520 particles tested
- EOL failure level of 1.28 x10^{-5} (upper 95% confidence limit)

Heatup tests in graphite furnace



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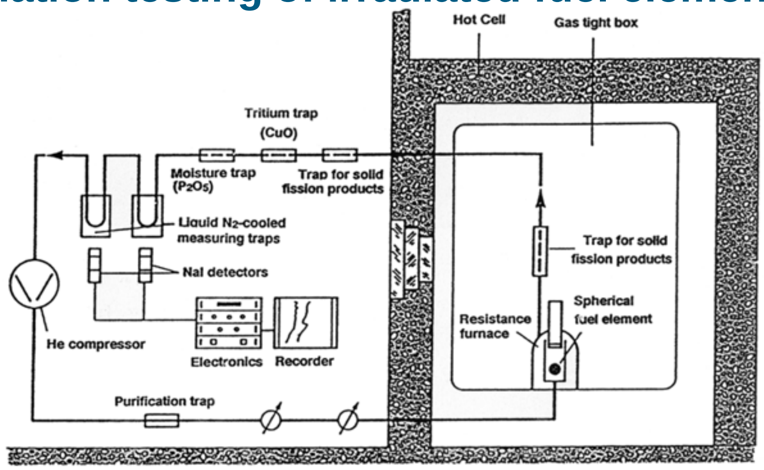
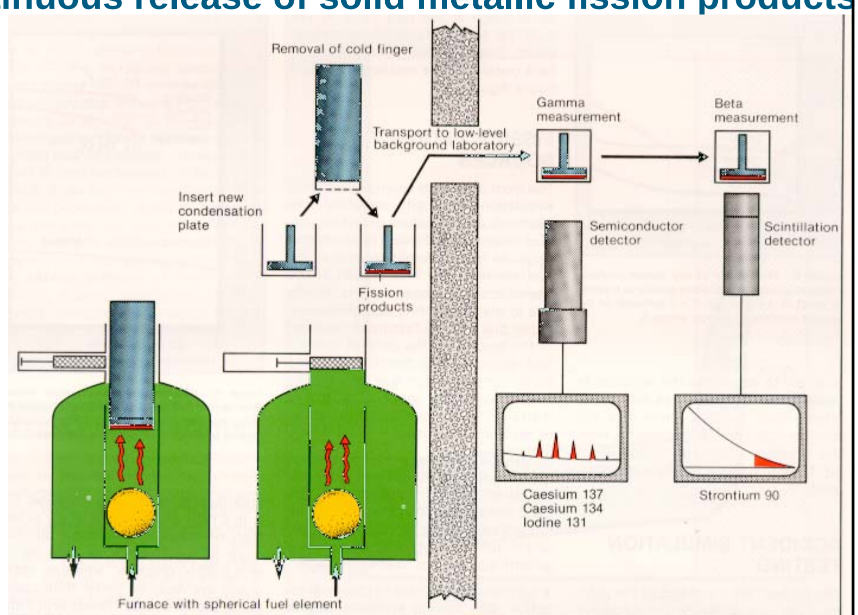
2.2 Observations from irradiation testing

- Advanced irradiation testing capabilities in Material Test Reactors (MTRs) have provided essential fuel particle performance data on a timely manner.
- In-reactor performance in real-time HTR environments and accelerated MTR environments are closely matched as expected provided no adverse affects occurred due to the accelerated neutron environment.
- Modern HTR TRISO fuel particles have been shown to retain fission products during normal operation and under accident conditions. Full demonstrations have been performed.

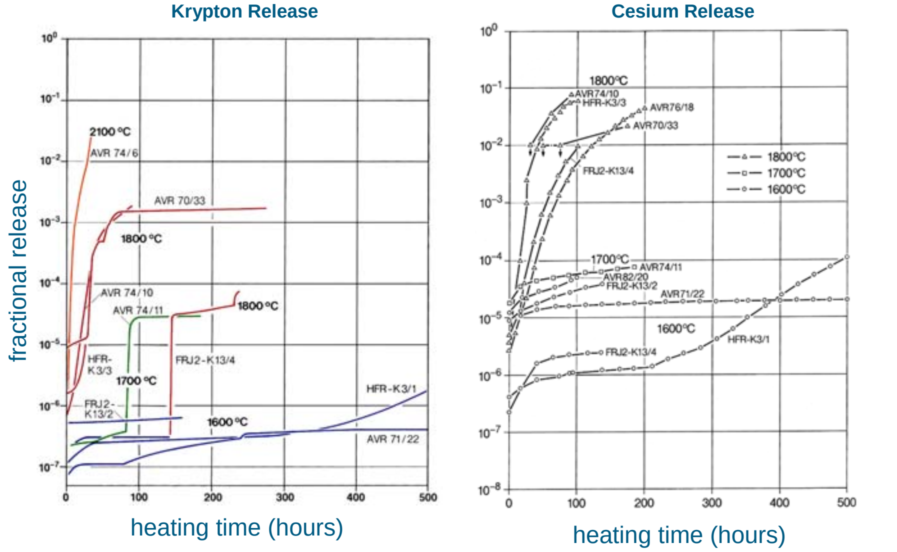
2.3 Cold finger apparatus (KÜFA) and associated instrumentation that monitors fission products during accident simulation testing of irradiated fuel elements



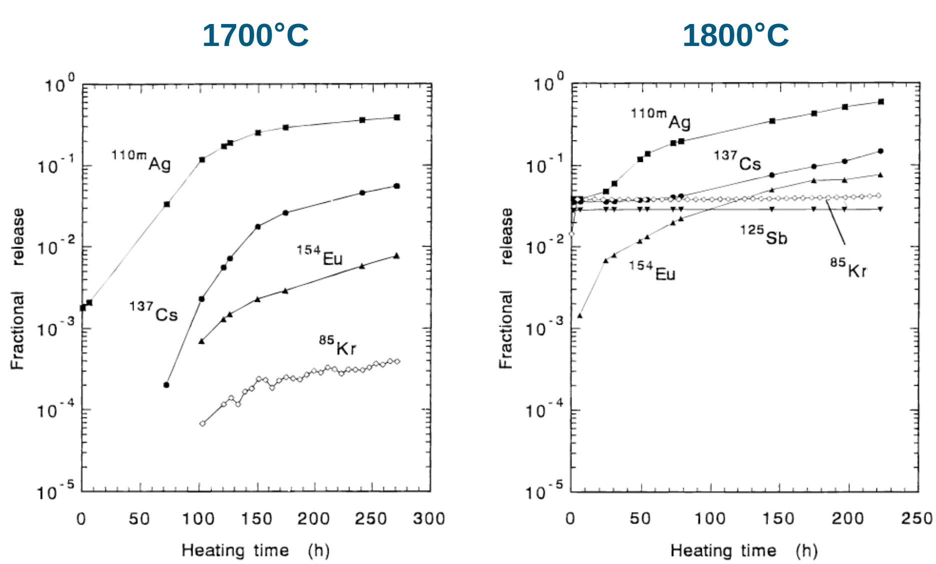
2.3 Accident conditions testing: quasi-continuous release of solid metallic fission products



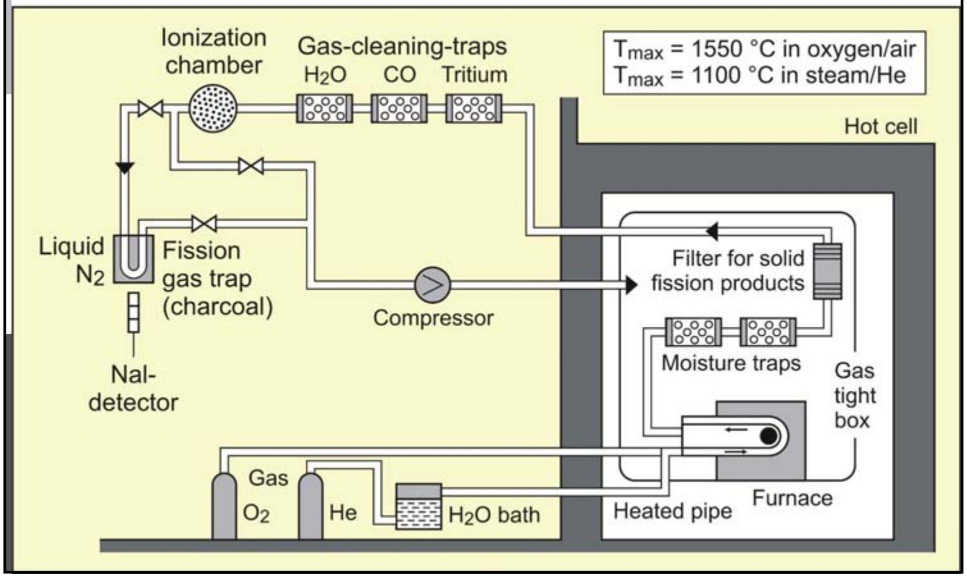
2.3 Jülich accident simulation testing with complete irradiation spherical fuel elements in the temperature range from 1600° to 2100°C



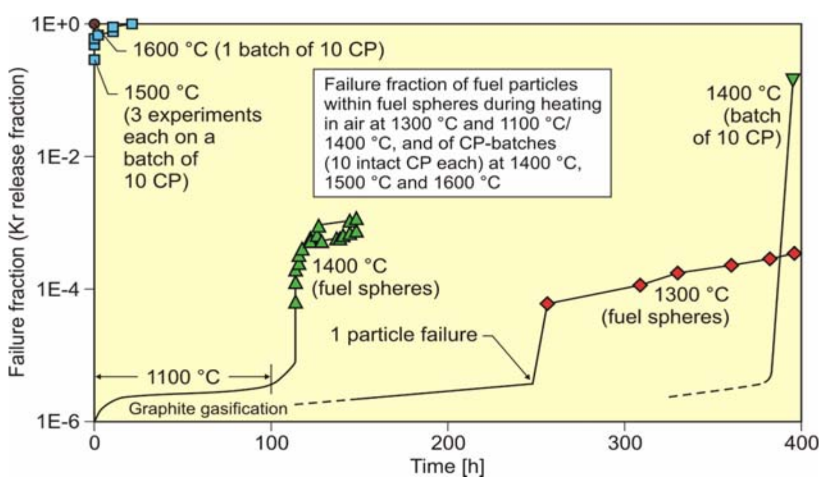
2.3 ORNL accident simulation testing Japanese coated particles irradiated in HRB-22 in the temperature range from 1700° to 1800°C source: K Minato et al, Nucl Techn 131 (2000), 36.



Scheme of the fuel element oxidation facility KORA

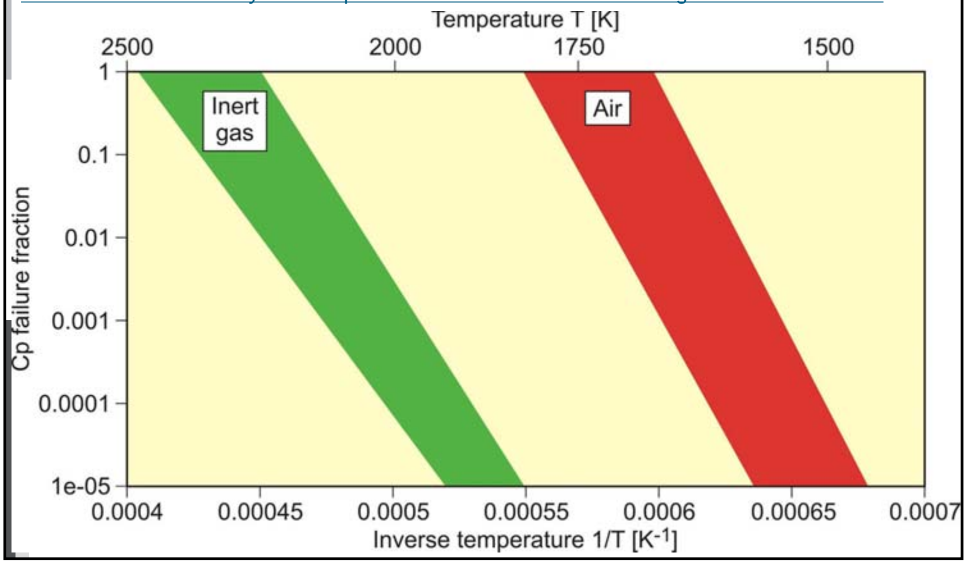


2.3 Destruction of TRISO particles in air – experimental results (KORA)



2.3 Estimated limits of TRISO particle performance: (left green): in inert atmospheres, (right red): in air.

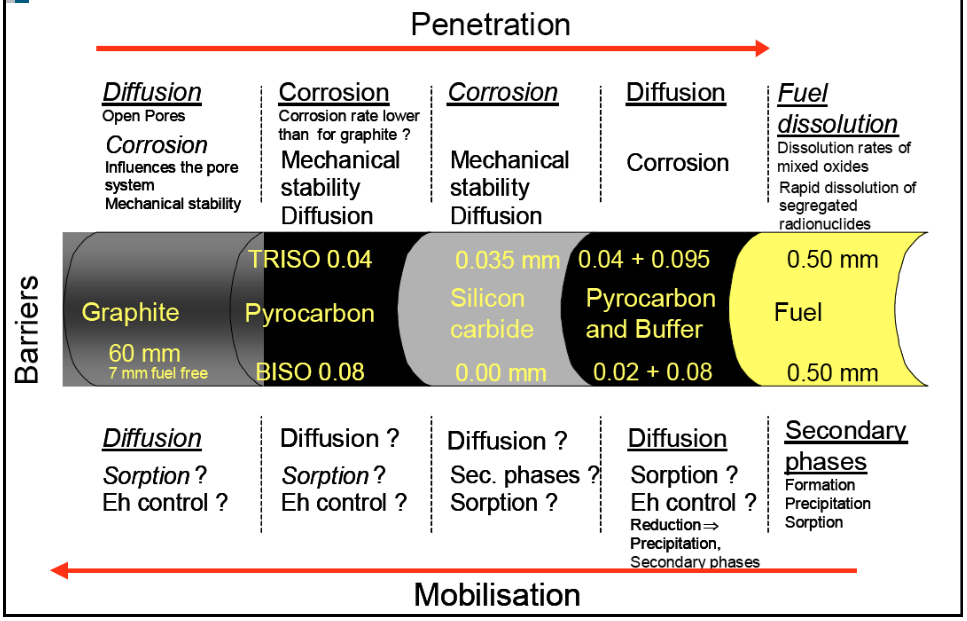
Data derived for a variety of burnups to 15% FIMA and various heating and oxidation times.



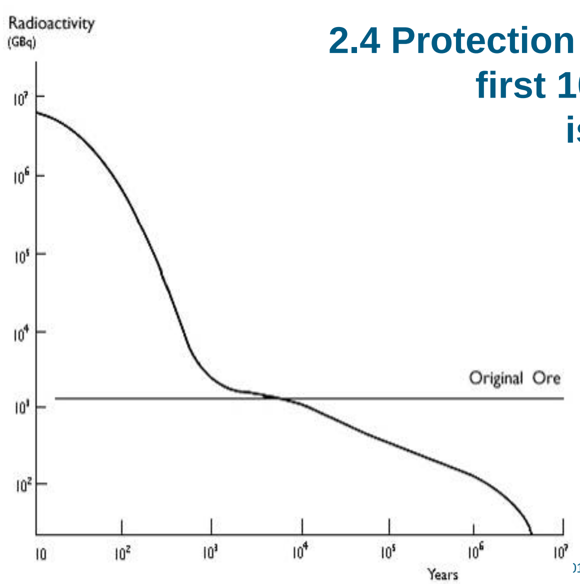
2.3 HTR fuel behavior in air under accident conditions:

- TRISO fuel is stable in air up to about 1300–1400°C; fast destruction at 1600°C
- Compared to He, failure temperatures in air are reduced by 400–600°C

2.4 Fuel disposal: proposed reaction mechanisms



2.4 Protection during the first 10,000 years is important



Fuel for Gas-Cooled Reactors, mainly HTR

3 Modeling and Performance Predictions

3.1 mechanical Failure

3.2 chemical failure

3.2 fission product transport

3.1 SiC properties

- 35 μm thick, $\pm$ 1.6 μm
- β-SiC, type 3C
- $\rho \geq 3.20$ g/cm^3 (practical theoretical density)
- CVD, $1500 \leq T_{dep} \leq 1600$°C, $v_{dep} \sim 0.2\mu$m/min
- columnar structure, but not too large grain size

3.2 SiC property data required

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- strength
  σ_0=834→687 MPa
  m = 8.0 →6.0
  unirrad. →irradiated at high temperatures

- fission product transport speed

$$D(Cs\_in\_SiC) = 6.7\times10^{-14}\left[m^2 s^{-1}\right]\ \exp\left(-\frac{125\,kJ/mol}{RT}\right)$$

$$D(Ag\_in\_SiC) = 3.6\times10^{-9}\left[m^2 s^{-1}\right]\ \exp\left(-\frac{215\,kJ/mol}{RT}\right)$$

- corrosion due to fp-SiC interaction
  resulting in thinning of layer thickness

$$\Delta v[m] = \int_0^t 5.87\times10^{-7}\left[ms^{-1}\right]\ \exp\left(-\frac{179.5\,kJ/mol}{RT(t')}\right)dt'$$

Pd attack limiting under accident conditions

- SiC thermal decomposition
  problem only at temperatures > 2000 ℃

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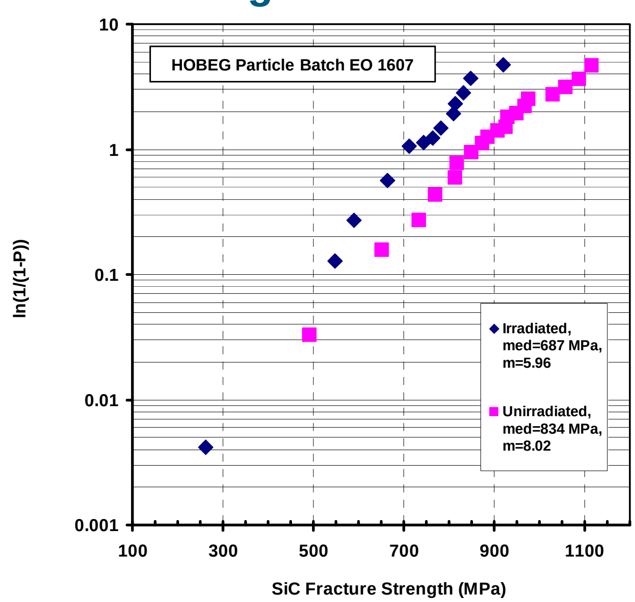
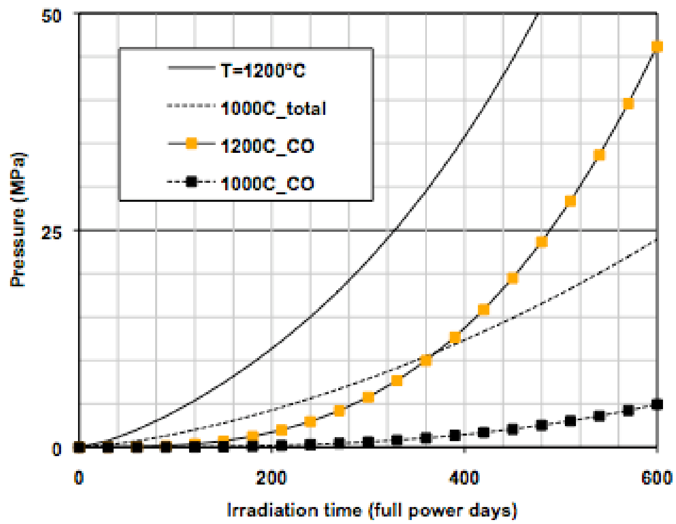
3.1 SiC strength



3.1 Gas pressure buildup during normal operation

Total pressure = fission gas Xe,Kr presure + CO pressure

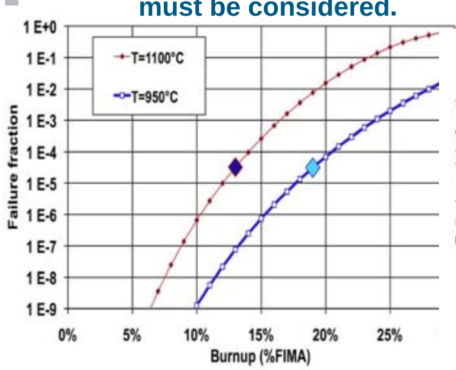


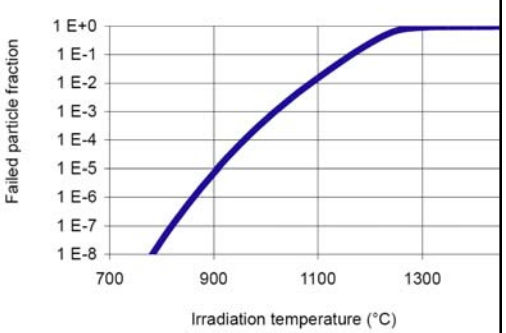
Nabielek slide 51

3.1 Mechanical coated particle failure

- internal gas pressure induces stresses in the coating layer
- when stress exceeds strength -> the layer fails, but
  - statistical strength distribution
  - statistical distributions of dimensions

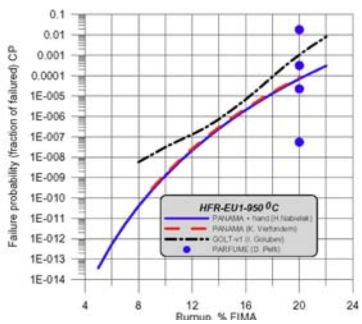
must be considered.

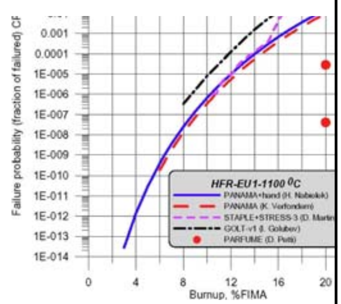




3.1 Mechanical coated particle failure

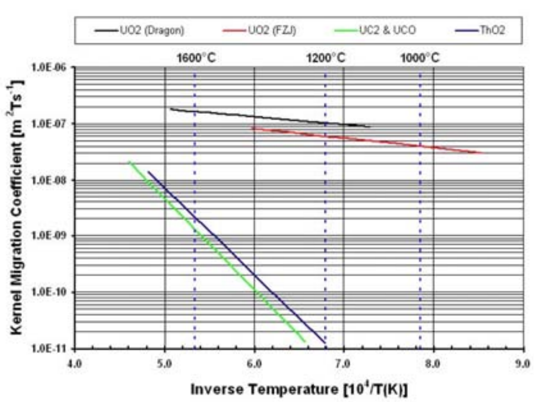
Prediction of coated particle failure probability for HFR-EU1 irradiation test with different computer models in international cooperation





3.2a Chemical failure: amoeba effect

- **UO_2 fuels only: kernel migration attacks coating layers**
- **particle fails when coating layer is getting too thin**
- **typically only in high power density fuel rods due to high temperature gradient**

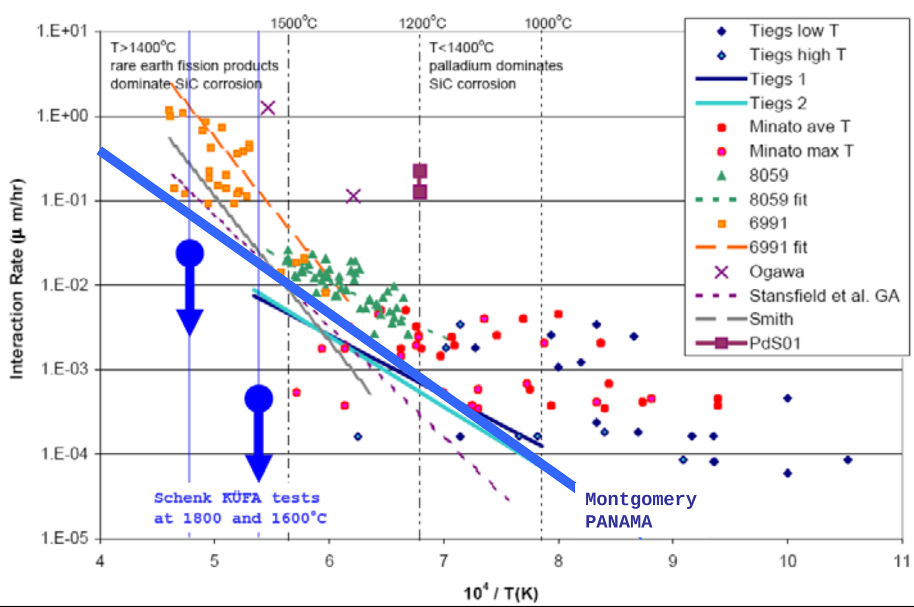


3.2a Chemical failure: Pd attack

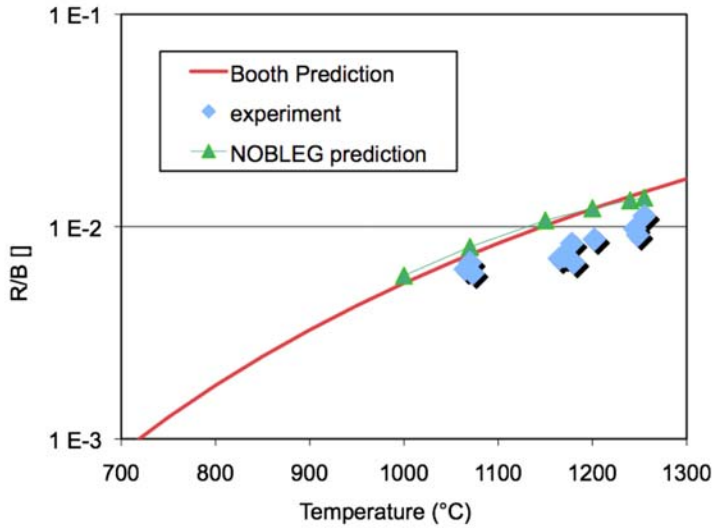
•chemical attack of fission products at the inner SiC surface

•particle fails when coating layer is getting too thin or too weak

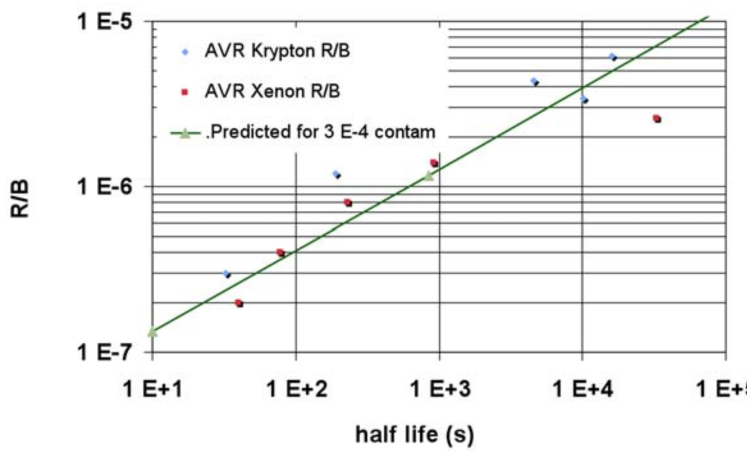
3.2a Chemical failure: Pd attack rates



3.3 Fission product transport, e.g. release prediction of short-lived fission gases in a test



3.3 Fission product transport, e.g. release prediction of short-lived fission gases in AVR



Conclusions on HTR fuel

- Modern HTR fuel particles retain fission products during normal operations and in accidents. Full demonstrations have been performed.
- Predictive models for mechanical particle performance and fission product transport are available.
- Safe retention and protection is being demonstrated for long-term storage conditions